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Nuclear Data for AFCI: Am and Np Cross-Section Evaluations

Summary

We have completed new evaluated nuclear data files for neutron reactions on 241Am and 237Np, for Advanced Fuel Cycle Initiative (AFCI) applications. Particular attention was paid to fission and capture reactions, where new experimental data, nuclear reaction modeling calculations, and statistical analyses of data sets led to improved cross-sections with reduced uncertainties. Our results are represented in the ENDF-6 format and have been processed using the NJOY code for use in transport, criticality, and transmutation calculations. We also describe nuclear model calculations and evaluations of proton and alpha particle emission from structural materials, for gas production and damage analyses.

New Evaluation of n+^{241}Am

This year, we have carried out a new evaluation of ^{241}Am neutron-induced reaction cross-sections. This isotope is part of the stream of nuclear waste produced in nuclear reactors and is expected to be significantly present in the composition of nuclear spent fuel to be burned in future accelerator-driven systems (ADS) or advanced reactors.

A recent uncertainty and sensitivity study by Palmiotti *et al.* showed that uncertainties on n+^{241}Am cross-sections contribute significantly to the total uncertainty on the multiplicative factor K_{eff} for an ADS-burner, with a typical fuel composition dominated by minor actinides (see Fig. 1). This new study supports our earlier priority list of minor actinides to be re-evaluated in light of the AFC Initiative.

Figure 9 : Selected Cross Section for K_{eff}

	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	tot
Pu9-σ_{fiss}	-	0.04	0.06	0.35	0.26	0.15	0.11	0.12	0.13	0.03	0.01	-	-	-	-	**0.51**
Pu1-σ_{fiss}	-	0.04	0.02	0.06	0.11	0.19	0.14	0.10	0.08	0.02	-	-	-	-	-	**0.30**
Np7-σ_{fiss}	0.02	0.17	0.25	0.62	0.10	0.01	-	-	-	-	-	-	-	-	-	**0.70**
Np7-ν	0.01	0.05	0.08	0.18	0.03	-	-	-	-	-	-	-	-	-	-	**0.21**
Am1-σ_{cap}	-	-	0.02	0.41	0.74	0.86	0.33	0.29	0.24	0.15	0.03	-	-	-	-	**1.32**
Am1-σ_{fiss}	0.04	0.41	0.61	0.84	0.07	0.02	0.01	0.01	0.01	0.01	-	-	-	-	-	**1.12**
Am1-ν	0.01	0.14	0.22	0.28	0.02	0.01	-	-	-	-	-	-	-	-	-	**0.38**
Am3-σ_{cap}	-	-	0.02	0.21	0.36	0.50	0.21	0.19	0.17	0.11	0.02	-	-	-	-	**0.74**
Am3-σ_{fiss}	0.02	0.23	0.33	0.44	0.02	0.01	-	-	-	-	-	-	-	-	-	**0.59**
Am3-σ_{inel}	-	0.12	0.18	0.50	0.18	0.20	0.02	-	-	-	-	-	-	-	-	**0.60**
Cm4-σ_{fiss}	0.03	0.26	0.35	0.99	0.11	0.03	0.02	0.02	-	0.01	-	-	-	-	-	**1.09**
Cm5-σ_{fiss}	-	0.03	0.05	0.16	0.19	0.19	0.16	0.14	0.12	0.07	0.01	-	-	-	-	**0.41**
Fe56-σ_{inel}	0.01	0.06	0.11	0.48	-	-	-	-	-	-	-	-	-	-	-	**0.49**
N15-σ_{el}	-	-	0.03	0.17	0.06	0.04	0.01	-	-	-	-	-	-	-	-	**0.19**
Pb-σ_{inel}	0.03	0.25	0.23	0.24	-	-	-	-	-	-	-	-	-	-	-	**0.41**
Pb-$\sigma_{n,2n}$	0.02	-	-	-	-	-	-	-	-	-	-	-	-	-	-	**0.02**
Bi-σ_{inel}	0.03	0.37	0.29	0.12	-	-	-	-	-	-	-	-	-	-	-	**0.49**
Bi-$\sigma_{n,2n}$	0.03	-	-	-	-	-	-	-	-	-	-	-	-	-	-	**0.03**

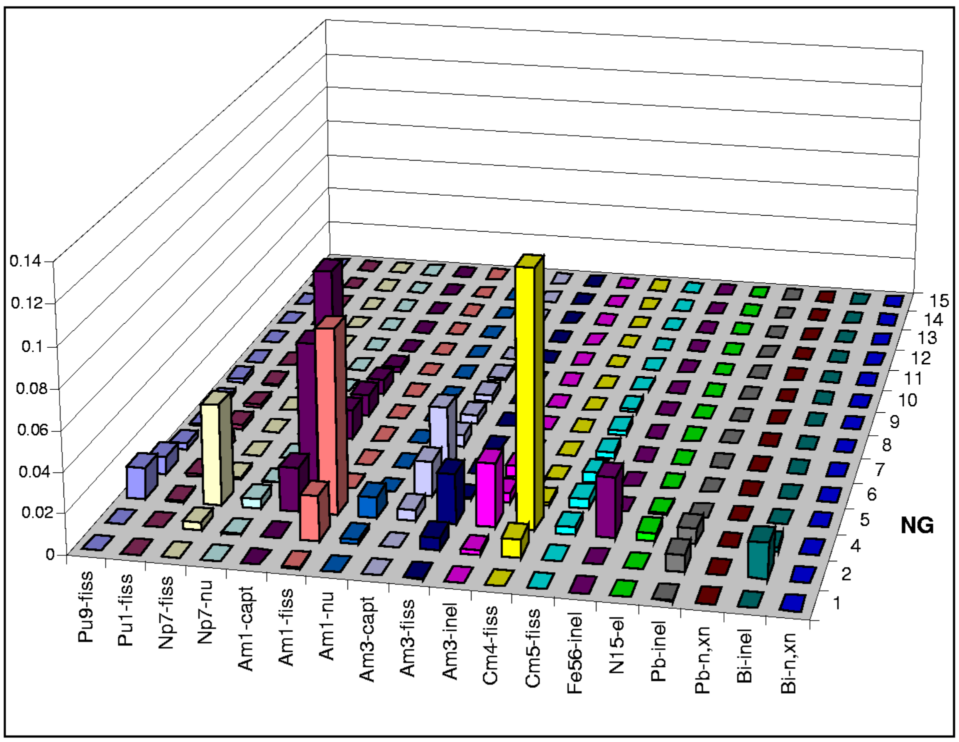

Fig. 1. Relative importance of selected cross-sections to the overall uncertainty on K_{eff} [from Palmiotti et al., ANL-AAA-036 report].

The latest ENDF/B-VI evaluation file for n+241Am dates back from 1994; since then, important new measurements became available, in particular for the (n,2n) reaction channel. The present work focused on new fission, capture, and (n,2n) cross-sections. The capture cross-section also includes information on the branching ratio to the metastable ^{242m}Am level ($T_{1/2}$=141y), which is important for estimating transmutation rates.

The ^{241}Am (n,f) cross-section was obtained via a generalized least-square approach to available experimental data. Measurements both absolute and in ratio to ^{235}U (n,f) cross-section were included in the analysis. Below 6.5 MeV, the old evaluated cross-section was unchanged, since results from our new measurement system were in very good agreement. Above 17 MeV, where experimental data are scarce, a GNASH nuclear model calculation was used to predict the (n,f) cross-section up to 30 MeV. The result is shown in Fig. 2.

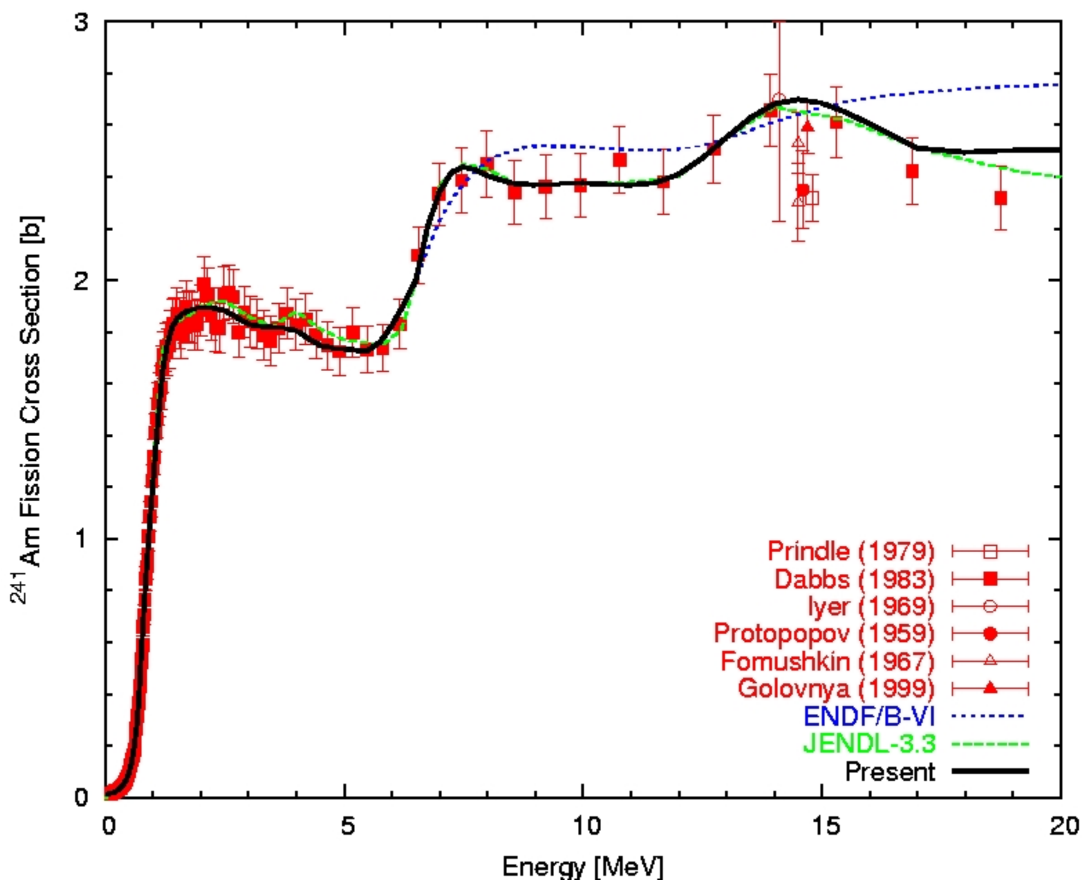

Fig. 2. Am-241 (n,f) cross-section.

GNASH nuclear model calculations were used to infer the (n,2n) and (n,3n) reaction cross-sections. Because of the large sensitivity of these channel cross-sections to the fission barrier parameters, these parameters were (simultaneously) adjusted to provide a best-fit to newly available experimental data. In particular, two differential experiments were used to guide this evaluation: Filatenkov (1999) and Lougheed (2001). In addition, integral data from LANL radiochemists were useful to guide the slope of the cross-section around 14 MeV. The result is depicted in Fig. 3. It definitely represents a large improvement over the previous ENDF/B-VI evaluation.

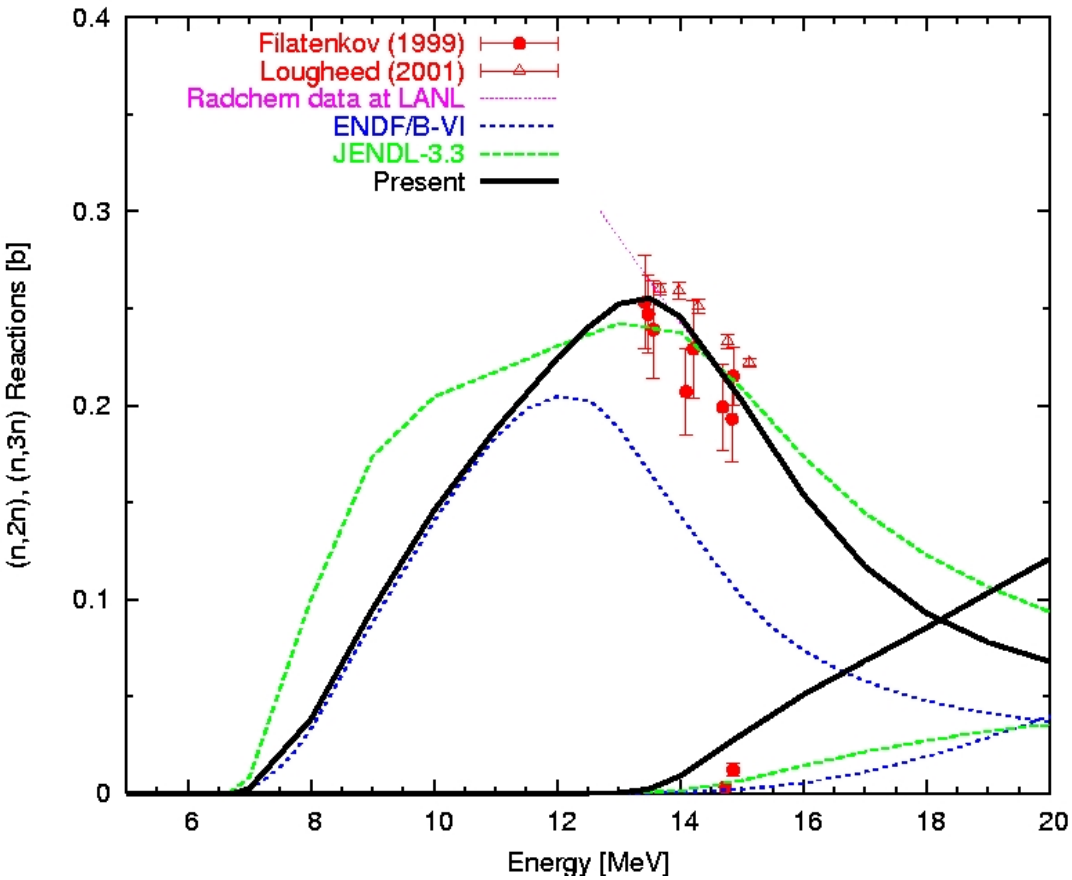

Fig. 3. Am-241 (n,2n) reaction cross-section.

Finally, the capture cross-section was also evaluated using GNASH predictions, and the branching ratio to the metastable state ^{242m}Am was renormalized to available experimental data. In particular, in the high-energy region the GNASH capture cross-section convoluted with the GODIVA neutron spectrum provided a way to assess this branching ratio and compare it to experimental integral data from GODIVA critical assembly. The final result is shown in Fig. 4.

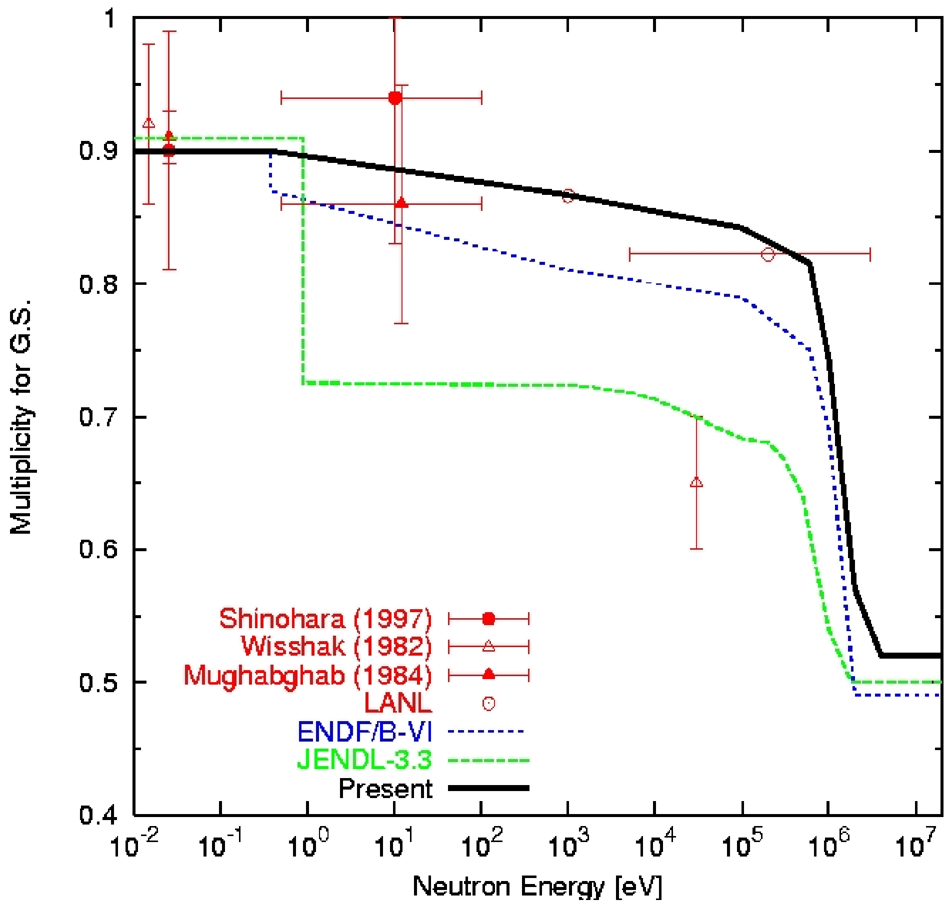

Fig. 4. Branching ratio to the metastable state in Am-242.

New Evaluation of n+^{237}Np

As shown in Fig. 1, uncertainties on the ^{237}Np (n,f) cross-section also contribute significantly to the total uncertainty on K_{eff}. The effect is particularly important in the fast-energy region. We have re-evaluated this cross-section in view of new experimental data by Lisowski, and a new assessment of the standard ^{235}U (n,f) cross-section in the 1—5MeV energy range.

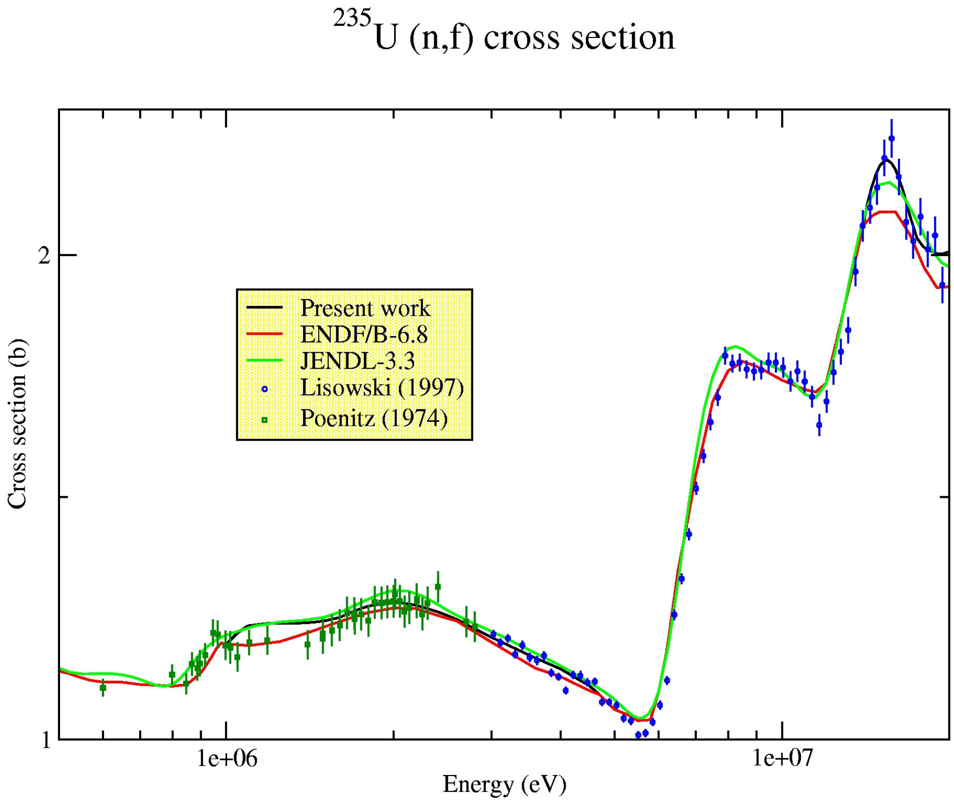

Fig. 5. New U-235 (n,f) cross-section.

In 2002, we re-evaluated the ^{239}Pu (n,f) cross-section for the AFC Initiative. In fact, most (n,f) cross-sections are measured in ratio to ^{235}U (n,f) cross-section, which is regarded as a standard in the ENDF/B-VI evaluation process. In particular, this means that this cross-section is thought to be known very precisely and is used to normalize other neutron cross-section measurements (such as ^{237}Np fission). However, systematic discrepancies among evaluations (JENDL-3.3 and ENDF/B-VI especially) and some clues from rare but valuable absolute measurements led us to re-evaluate this cross-section. The result is shown in Fig. 5. Our new evaluation tends to lay right between JENDL-3.3 and ENDF/B-VI curves. This new cross-section is now being supported by a very recent (and still preliminary) evaluation by A. Carlson and V. Pronyaev, who are working toward the release of ENDF/B-VII standards.

Since most experimental data for ^{237}Np (n,f) cross-section are in ratio to the ^{235}U (n,f) cross-section, our new evaluation for ^{237}Np (n,f) reflects the above mentioned changes. Figure 6 shows the new cross-section as compared to the older evaluated ENDF/B-VI.

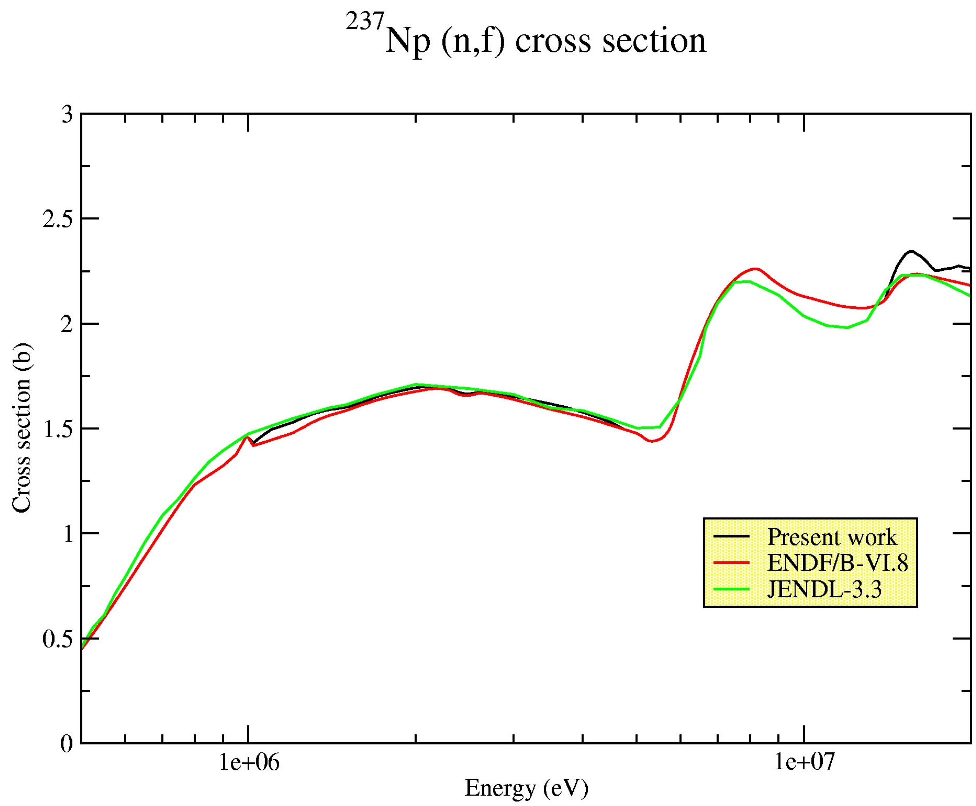

Fig. 6. New Np-237 (n,f) cross-section vs. ENDF/B-VI.8 and JENDL-3.3 evaluations.

We also studied other important cross-sections in ^{237}Np to determine if they are in need of upgrading. Our conclusion was that the evaluated data in the current ENDF/B-VI file (from an earlier Los Alamos evaluation) does not need to be modified at this time. The nu-bar evaluation (average number of prompt fission neutrons) and the inelastic scattering cross-sections appear to be very reasonable. This is important since an NEA Working Party on Evaluation Cooperation (WPEC) document highlighted some significant discrepancies in ADS criticality using different nuclear data sets from different countries (US vs. Japan), and we traced this discrepancy to different evaluations for ^{237}Np nuclear data, particularly nu-bar. Our present study has shown that the US ENDF evaluation for nu-bar is more reliable than the Japanese JENDL-3.2 evaluation, and indeed the newly released JENDL-3.3 evaluation for neptunium now better agrees with our ENDF evaluation.

Data Testing

We have incorporated these changes into an ENDF file and have performed integral data testing in LANL fast critical assemblies by comparing the ^{237}Np/^{235}U fission ratio in

Jezebel, Godiva, Jezebel-23 and Bigten & Flattop. Experiment and theory for the ^{237}Np to ^{235}U fission ratio agree to within 4%—5%. However, we believe that the neptunium cross-section itself is accurate to 2%—3%, and the larger uncertainties (4%—5%) come from modeling deficiencies in our calculated neutron spectrum in the assembly. This is because ^{237}Np and another threshold fissioner, ^{238}U, would have an underpredicted fission cross-section ratio to ^{235}U fission if the calculated spectrum is a little too soft.

Results for new actinide evaluations: Note the ^{237}Np/^{235}U fission ratio in the assembly is denoted by "37/25."

Uranium-235 assembly

CSEWG Godiva
- k_{eff}= 0.99934(21)
- 28/25 = 0.9606
- 37/25 = 0.9540
- 23/25 = 0.9871
- 39/25 = 0.9742

CSEWG Flattop-25
- k_{eff} = 1.00236(25)
- 28/25 = 0.9701
- 37/25 = 0.9712
- 23/25 = 0.9747
- 39/25 = 0.9797

HEU-MET-FAST-001 (Godiva)
- k_{eff} = 0.99888(21)
- 28/25 = 0.9571
- 37/25 = 0.9561
- 23/25 = 0.9876
- 39/25 = 0.9750

HEU-MET-FAST-028 (Flattop-25)
- k_{eff} = .99791(23)
- 28/25 = 0.9714
- 37/25 = 0.9728
- 23/25 = 0.9746
- 39/25 = 0.9800

Plutonium-239 assembly

CSEWG Jezebel
- k_{eff} = 1.00037(21)
- 28/25 = 0.9767
- 37/25 = 0.9720
- 23/25 = 0.9885
- 39/25 = 0.9731

CSEWG Flattop-Pu
- k_{eff} = 1.00189(25)
- 28/25 = 0.9788
- 37/25 = 0.9762

PU-MET-FAST-001 (Jezebel)
- k_{eff} = 0.99983(21)
- 28/25 = 0.9768
- 37/25 = 0.9744
- 23/25 = 0.9886
- 39/25 = 0.9738

PU-MET-FAST-006 (Flattop-Pu)
- k_{eff} = 1.00042(25)
- 28/25 = 0.9827
- 37/25 = 0.9810

Uranium-233 assembly

CSEWG Jezebel-23
- k_{eff} = 0.99872(21)
- 28/25 = 0.9810
- 37/25 = 0.9805

CSEWG Flattop-23
- k_{eff} = 0.99988(24)
- 28/25 = 0.9778
- 37/25 = 0.9818

U233-MET-FAST-001 (Jez23)
k_{eff} = 0.99845(20)
28/25= 0.9858
37/25= 0.9820

U233-MET-FAST-006 (Flat23)
k_{eff} = 0.99837(25)
28/25 = 0.9698
37/25 = 0.9781

CSEWG 1-D Bigten
k_{eff} = .99452(18)
(exp=.996)
28/25 = 0.9614
37/25 = 0.9429
23/25 = 0.9712
39/25 = 0.9727
8c/25 = 0.9725

IEU-MET-FAST-007 (2-D Bigten)
k_{eff} = 0.99211(18)

28/25 = 0.9526
37/25 = 0.9383
23/25 = 0.9713
39/25 = 0.9721
8c/25 = 0.9738

Notation:
28/25 is U238f/U235f
37/25 is Np237f/U235f
23/25 is U233f/U235f
39/25 is Pu239f/U235f
8c/25 is U238c/U235f

We have also performed integral data testing of our new ^{237}Np ENDF evaluation by simulating the recently constructed composite Np-U fast criticality at the Los Alamos LACEF facility. Agreement between calculation and measurement for the k_{eff} is reasonable (about 0.993). Future work would be needed to determine whether agreement can be brought to the 1.00 level through nuclear data improvements (both in the ^{237}Np core and in the surrounding ^{235}U driver). However, LACEF experimentalists are still refining their MCNP model of this Np-U critical system, and we are unable to further address these issues until the model is finalized.

Gas Production in Structural Materials

We have also undertaken FY03 work on analyzing new LANSCE measurements for gas-production, specifically hydrogen and alpha particle production. Our LA150 cross-sections that we produced a few years ago, that are widely used in ADS and AFC applications, include information on these cross-sections and spectra. The new measurements extend to higher energies (~100 MeV). We have performed GNASH nuclear model calculations to compare against these data — and we see a need to upgrade nuclear level densities to better represent the measurements. Once the LANSCE data are finalized, we will upgrade the LA150 evaluations.

Documentation of ^{239}Pu evaluation

We have made progress in documenting our earlier LA150 ^{239}Pu evaluation for AFC in a refereed journal article, to be submitted to *Physical Review C*. The article describes the Lisowski LANSCE fission measurements and the nuclear theory to interpret these measurements.